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RECENT DEPOSITION OF ^{210}Pb ON THE GREENLAND ICE SHEET: VARIATIONS IN SPACE AND TIME

by

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ABSTRACT

Detailed ^{210}Pb profiles were determined for four "Chernobyl dated" snowpits sampled during a wide-ranging survey of the Greenland ice sheet during the 1988 season. The profiles from widely separated pits show little or no coherence; even for two pits only 40 km apart the profiles differ in detail. There does not appear to have been any seasonality in the deposition of ^{210}Pb onto the ice sheet in the two years since the Chernobyl accident. The total deposition of ^{210}Pb during this period ($10\text{--}20\text{ bq m}^{-2}$) was about 20 times less than has been observed at mid-latitude sites in the eastern United States. The three pits west of the ice-sheet divide recorded very similar depositional fluxes, while the one eastern pit had only two-thirds the average of the others, suggesting a west-to-east gradient in the deposition of ^{210}Pb , and perhaps other continentally-derived submicron aerosols, onto the Greenland ice sheet.

INTRODUCTION

Glacial ice from around the world is increasingly regarded as representing perhaps the most important archive of information on past climate and atmospheric conditions. Because of this interest, the number, diversity and quality of records recovered from glacial snow and ice is rapidly increasing. However, in order to decipher the information contained in the depth profiles of the various parameters measured in ice, two key points must be addressed: the conversion of depth in the glacier to a temporal framework, and the relation between the concentrations measured in ice and those in the atmosphere at the time the snow fell. Atmospheric radionuclides can contribute greatly in both of these areas.

A substantial body of research has clearly established the utility of natural (mainly ^{210}Pb) and anthropogenic (derived from bomb testing) atmospheric radionuclides for dating glacial snow and ice (Goldberg, 1963; Crozaz and others, 1964; Picciotto and others, 1964, 1967; Crozaz and Langway, 1966; Jouzel and others, 1979; Gunten and others, 1982; Gäggeler and others, 1983). The relatively high accumulation rates on the Greenland ice sheet limit the application of these dating techniques to studies where samples are taken from depths (10s to 100s of m) much greater than those to which snowpits can routinely be dug. The nuclear reactor accident at Chernobyl in April 1986 injected enough radioactive material into the atmosphere to provide a valuable new time horizon in the upper layers of the snow on the Greenland ice sheet (Dibb, 1989).

Atmospheric ^{210}Pb results from the decay of the gaseous, ^{238}U decay series radionuclide, ^{222}Rn emitted from continents (Turekian and others, 1977). Very shortly after production the ^{210}Pb associates with submicron aerosols (Bondietti and others, 1987, 1988) and thereafter shares the same fate as the aerosol. As a result, atmospheric ^{210}Pb serves as a tracer of continentally-derived submicron aerosols in the atmosphere.

In Antarctica several long-term studies have monitored aerosol concentrations of ^{210}Pb to help clarify the atmospheric transport processes bringing aerosols and their associated chemical species to the ice sheet (Lockhart and others, 1966; Maenhaut and others, 1979; Wagenbach and others, 1988). Although not conclusive, the results have suggested the importance of stratospheric transport in bringing continentally-derived aerosols to high southern latitudes. In the Arctic, Rahn and McCaffrey (1980) and Daisey and others (1981) observed a pronounced seasonality in ^{210}Pb aerosol concentrations at Barrow (winter maximum with concentrations 15 times higher than in summer) that was corroborated by Graustein and Barrie (personal communication from W. Graustein) during a study at Mold Bay. The seasonal pattern of ^{210}Pb in the aerosol at these high-latitude, low-altitude sites was very similar to the seasonality of the pollution phenomenon known as Arctic Haze; in particular, Rahn and others (1980) reported very high correlations between the concentrations of ^{210}Pb and SO_4^{2-} . They proposed that the annual migration of the Polar Front from its high-latitude summer position to a more southern winter position brought large, mid-latitude, continental areas effectively into the polar air mass, so that ^{210}Pb and pollutants emitted from these areas in the winter are readily transported to high latitudes.

The present study describes detailed ^{210}Pb profiles from the top 3 m of four "Chernobyl dated" snowpits on the Greenland ice sheet. The primary goals of the investigation were to ascertain whether ^{210}Pb might provide a strong seasonal signal to assist in the interpretation of detailed chemical records recovered from these and future shallow pits in Greenland, and to examine spatial and short-term temporal variability in the deposition of ^{210}Pb on the Greenland ice sheet.

METHODS AND MATERIALS

Large volume (approximately 1 litre when melted) snow samples for radionuclide analyses were collected in a series of snowpits in Greenland as part of a regional snow chemistry survey conducted in 1987 and 1988 (Fig. 1). In relation to the overall survey, the primary purpose of the radionuclide sampling was to determine the distribution of fallout on the Greenland ice sheet from the Chernobyl reactor accident. To achieve this goal 2 m sections of seven snowpits, centered on what the field party estimated was the level of the 1986 summer surface, were sampled at 6 cm intervals. In four of the pits dug in 1988 the entire depth was sampled in order to examine in detail the recent history of ^{210}Pb deposition. All of the samples were immediately double-bagged in polyethylene and returned to New Hampshire frozen, where they were stored at -20°C until preparation for analysis.

Upon removal from the freezer the outside 0.5 cm of the originally vertical sides of each block were carefully scraped off, then concentrated HCl (0.333 ml kg^{-1} sample) and ^{208}Po tracer (for the samples destined for ^{210}Pb

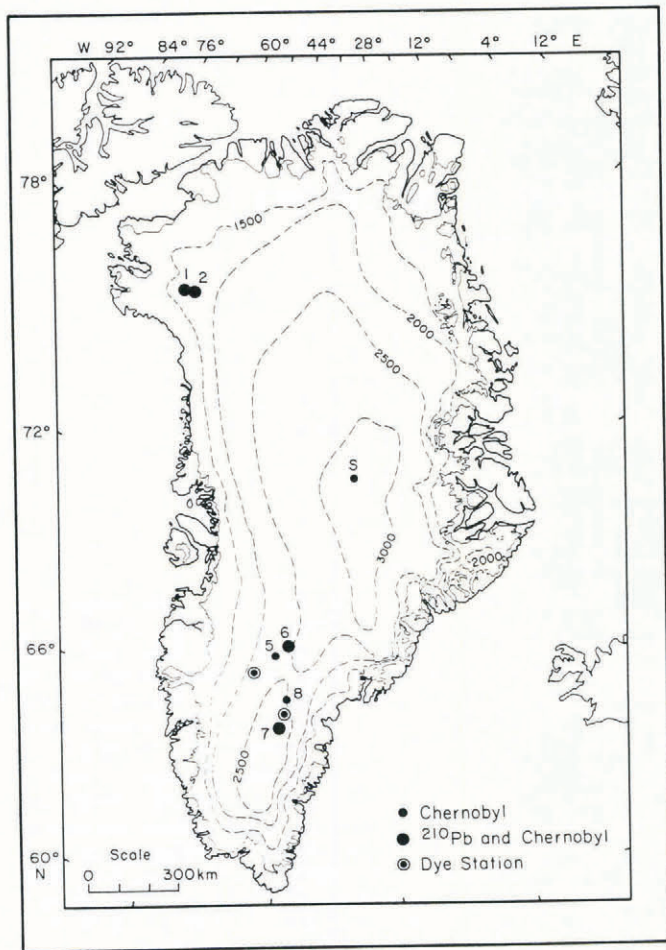


Fig. 1. Location of pits where large-volume samples were collected for radionuclide analyses. The Summit (S) pit was sampled in the 1987 season and the rest were sampled during the 1988 season.

analysis) were added to the samples. The radionuclides were concentrated from the melted samples onto cation exchange filters (Delmas and Pourchet, 1977). Each sample was filtered three times, the filters dried and then placed in polyethylene vials for gamma spectrometric determination of ^{134}Cs and ^{137}Cs . After gamma counting the filters were placed in a porcelain funnel and leached with 5 ml of concentrated HCl. The leachate was collected in acid-washed teflon beakers, to which 20 ml of Milli-Q deionized H_2O and 5 ml of Na-citrate was added. Polonium was then spontaneously plated onto polished silver planchets, for four hours at 70°C with frequent gentle swirling and then overnight at room temperature. ^{210}Pb activities were determined from the ratio of the alpha activities of the daughter ^{210}Po and the tracer ^{208}Po (Crozas and Fabri, 1966; Flynn, 1968).

RESULTS AND DISCUSSION

Fallout from Chernobyl was found in all of the pits. The distribution of this radioactive debris on the Greenland ice sheet is discussed more fully elsewhere (Dibb, 1989). However, the presence of a layer of snow labeled with Chernobyl fallout in each of the pits does provide a very well constrained temporal framework in which to consider the ^{210}Pb results.

The depth profiles of ^{210}Pb concentrations in the top 3 m of the four pits are noisy (Figs 2 and 3). Perhaps the most striking aspect of these profiles is the lack of any obvious seasonality in the concentration of ^{210}Pb in recent Greenland snow, considering the previously mentioned 15-fold increase in ^{210}Pb concentration observed in the aerosol at Barrow during the winter (Rahn and McCaffrey, 1980; Daisey and others, 1981) and the 21-fold increase observed by Graustein and Barrie at Mold Bay (personal

communication from W. Graustein). Davidson and others (1985, 1989) have suggested that the seasonal pattern of SO_4^{2-} deposition in snow at Dye 3 follows the annual cycle observed for aerosol concentrations at a number of stations around the Arctic (winter maximum and summer minimum), but that the signal is greatly dampened in the snow due to the prevalence of rimed snow (with much higher scavenging efficiency) occurring during the summer. Such a mechanism might explain the apparent lack of a winter peak in ^{210}Pb concentration in the snow falling on the Greenland ice sheet in recent years. Alternatively, it may be that the presence of the Greenland ice sheet has enough of an influence on the atmospheric dynamics around and above it that the climatology of ^{210}Pb (and perhaps other continentally derived submicron aerosol-associated species) over the ice sheet has little or no relation to what has been described for other regions of the Arctic. (A 12-month aerosol sampling campaign at Dye 3 did not reveal any pronounced increase in the concentration of ^{210}Pb during the winter of 1988-89 (Dibb, in preparation).)

The details of the ^{210}Pb profiles show little coherence between the three widely separated pits (Fig. 2). Pits 1 and 2, separated by 40 km, have ^{210}Pb profiles that are more

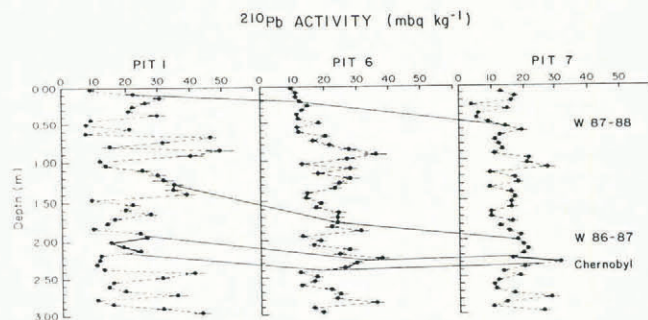


Fig. 2. Detailed profiles of ^{210}Pb activity versus depth in the firn for pits 1, 6 and 7. Pits were sampled at continuous 0.06 m intervals. The horizontal bars represent one sigma counting uncertainty. The depth of the winter layers was derived from $\delta^{18}\text{O}$ profiles (not shown). The Chernobyl layer is from Dibb (1989).

similar but still differ in detail (Fig. 3). This is not surprising, considering the importance of wet deposition processes in carrying aerosol-associated species to the surface of the ice sheet (Junge, 1977; Davidson, 1989) and the extreme heterogeneity of these processes in space and time. However, if longer intervals of time are examined, some interesting trends appear.

Over the two years since the deposition of the Chernobyl debris layer, the average concentration of ^{210}Pb in snow has been 23.3, 21.8, 20.2 and 14.8 mbq kg^{-1} at the locations of pits 1, 2, 6 and 7 respectively. Given the depth to the Chernobyl layer (2.04, 2.28, 2.34 and 2.28 m,

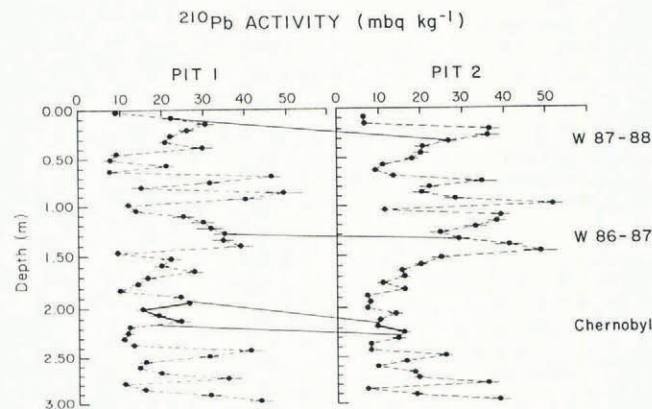


Fig. 3. Detailed profiles of ^{210}Pb activity versus depth in the firn for pits 1 and 2. Pits were sampled at continuous 0.06 m intervals. The horizontal bars represent one sigma counting uncertainty. The depth of the winter layers was derived from $\delta^{18}\text{O}$ profiles (not shown). The Chernobyl layer is from Dibb (1989).

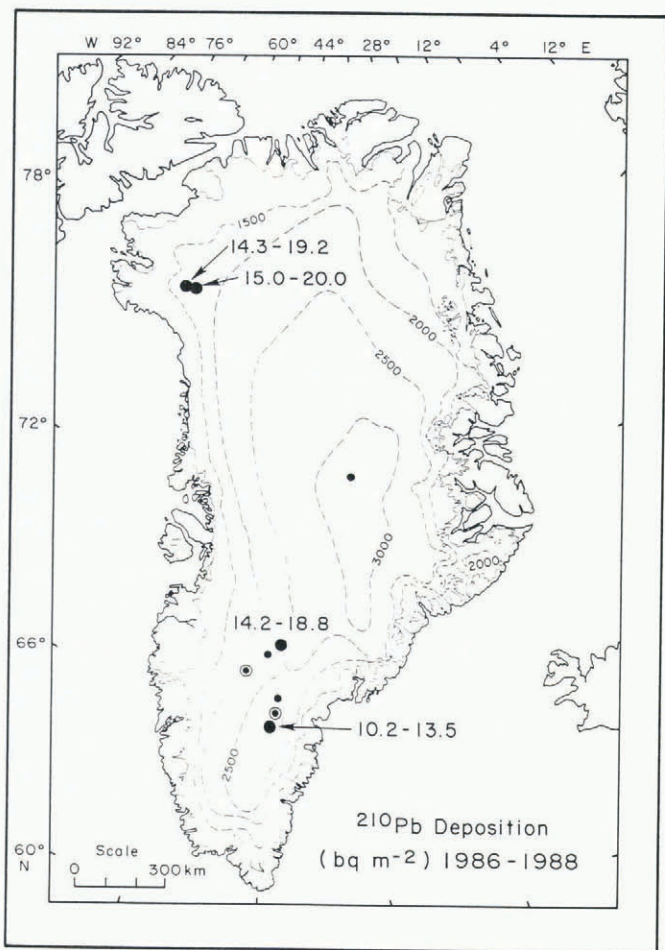


Fig. 4. ^{210}Pb deposition at the four sites during the two-year interval between the Chernobyl accident in late April, 1986, and sampling in May, 1988. The range in estimated deposition reflects the uncertainty in the density of the firn over the depth sampled. The low estimate for each pit assumes an average density of 300 kg m^{-3} and the high estimate assumes 400 kg m^{-3} .

respectively), and assuming an average density in the range of $300\text{--}400\text{ kg m}^{-3}$, the total amount of ^{210}Pb deposited during that two-year period has been calculated for each of the pits (Fig. 4). Even the highest estimate of ^{210}Pb deposition onto the Greenland ice sheet (20 bq m^{-2} in two years) is almost 20 times lower than those observed at continental sites. Olsen and others (1985) measured fluxes of 152 and $173\text{ bq m}^{-2}\text{ y}^{-1}$ at sites in Virginia and Tennessee that were very similar to the 5-year average of $177\text{ bq m}^{-2}\text{ y}^{-1}$ reported by Turekian (1983) for a coastal Connecticut site. The lower ^{210}Pb flux to the surface of the Greenland ice sheet reflects both loss of submicron aerosols during the long transport from the continental source areas and the limited amount of precipitation onto the ice sheet.

The very close agreement in the ^{210}Pb fluxes recorded in pits 1, 2 and 6 is striking, given the aforementioned lack of correspondence between the detailed profiles (Figs 2-4). The flux at pit 7 is only about two-thirds that at the other pits. It is interesting that pit 7 is the only one of the four that is east of the ice-sheet divide. Pit 7 is also further south and closer to the divide than the other pits, but the suggestion of a significant difference in ^{210}Pb deposition on either side of the divide is intriguing. The proximity of North America, particularly Arctic Canada, to the west coast of Greenland and the long transport pathways over water from potential Eurasian source areas suggest North America as perhaps the dominant source of ^{210}Pb in Greenland snow. The apparent west-to-east gradient in ^{210}Pb deposition might then reflect progressive removal of ^{210}Pb (and, by implication, other continentally derived submicron aerosols) as air masses cross Greenland from the west. Obviously, four pits constitute a very sparse data set and the dynamic interactions between transport and

deposition processes in the atmosphere are complex; however, it is interesting to note that Heidam (1984) observed higher concentrations of crustally derived species during 1979-80 at a station on the west coast of Greenland (GOVN) than at two stations on the east coast (NORD and KATO).

CONCLUSIONS

The natural atmospheric radionuclide ^{210}Pb , as it is preserved in snow and firn deposits, has great potential as a tracer of atmospheric transport and deposition processes in addition to its well established role as a dating tool. The lack of seasonality in ^{210}Pb deposition on the Greenland ice sheet suggests that the air masses over Greenland are decoupled to some extent from the general high Arctic atmosphere. On very short time scales, ^{210}Pb indicates the spatial heterogeneity of the deposition (and subsequent preservation) of submicron aerosols on the Greenland ice sheet. For periods on the order of two years (perhaps as short as one year) ^{210}Pb deposition appears to be nearly uniform over large regions of the ice sheet. The very limited data set described here suggests significantly more ^{210}Pb deposition west compared to east of the ice-sheet divide, which agrees with previous work showing higher concentrations of crustally-derived species in the aerosol on the west than the east coast of Greenland. Similar studies, particularly to verify the gradient in deposition hinted at by the present results, should further our understanding of the linkages between snow and ice chemistry records and the atmospheric processes that are responsible for these records.

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